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MATHEMATICAL MODELING OF RADIOACTIVE WASTE GLASS MELTER (U)

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MATHEMATICAL MODELING OF RADIOACTIVE WASTE GLASS MELTER

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ABSTRACT

The radioactive waste glass melter used at Savannah River Site (SRS) is a liquid slurry feed joule-heated ceramic melter. The physical nature of a joule-heated meter is complex and involves interactions between electric, thermal, and flow fields. These interactions take place through strongly temperature-dependent glass properties, natural convection, advection, diffusion, and volumetrically distributed joule heating sources. The cold feed on top of heated glass distabilizes the flow field and develops unsteady asymmetric flow motions underneath. Thus waste glass modeling requires solving a full 3-D, unsteady, momentum, energy, and electric equation with temperature-dependent properties. Simulation of noble metal deposit process requires an additional mass diffusion equation that is coupled to the momentum equation through mass advection term. The objective of this paper is to identify critical issues anticipated in the Defense Waste Process Facility (DWPF) melter operation and address how these issues can be resolved with current state-of-the-art mathematical modeling techniques.

INTRODUCTION

The past twenty four years have seen the high level radioactive waste vitrification process evolving from a bench scale operation to today's high-capacity process. Over this period, the process changed from a batch process where the feed is directly heated to melting temperature in a canister to the current continuous slurry feed process in which the feed is melted through joule-heating process in a melter and then delivered to a canister [1]. In the joule-heating process, the glass acts as electrolyte at high temperatures and generates heat when electric currents are passed through. In the absense of active mixing devices, convection currents are the principal means of mixing in a melter and need to be promoted. Enhanced convection currents, however, increase refractory erosion rates and has a potential to shorten melter life. Therefore balancing glass mixing efficiency against refractory erosion rate is the key to a successful melter operation.

The melter mixing efficiency depends on the interactions between glass temperature, velocity, and electric field in the melt pool. Mathematical modeling provides a cost effective means of identifying these interactions. These interactions are governed primarily by the design parameters such as melter dimensions and electrode locations and secondarily by the operation parameters such as heater control, power skewing, and melt rate. Due to the high cost and non-modular nature of design, the melter design parameters are difficult to change. On the other hand, melter operation parameters are difficult to characterize by conventional experimental means. Therefore the need to apply mathematical modeling techniques as alternative numerical experiment tools has become more crucial than ever.

A critical operational issue that drew notable attention in recent years involves noble metal deposit process. Noble metals or nickel sulfides in the melter feed are heavier and have lower solubility than other chemical species. In reducing conditions, these metals amalgamate and tend to settle on the melter floor with potential to short-circuit the melter. A mathematical model is currently being developed at SRS to characterize this noble metal deposit process. This model is essentially based on mass diffusion equation which is coupled to the glass convection model that solves a full 3-D, unsteady, momentum, energy, and electric equation. The computer code currently being used for glass convection model at SRS is a modified version of FIDAP™. The present paper discusses mathematical modeling techniques required to resolve melter operation issues with a primary focus on noble metal deposit problem.

MELTER SYSTEM MODEL

The DWPF melter is a large scale liquid feed joule heated ceramic melter [2] (Figure 1). As 35 percent of the total volume and 70 percent (10^9 Ci) of the radioactivity of all Defense High-Level Waste (DHLW) accumulated in the U.S. is stored at SRS, the DWPF vitrification process is considered to be a reference process. The DWPF melter takes slurry form of radioactive fission products mixed with borosilicate glass and water. The water in the slurry vaporizes immediately and the solid portion of the waste continuously piles up and melts at the same time, forming a floating layer called "cold cap". The cold cap is melted underneath by two diametrically-opposed plate electrodes. The melted glass is delivered through the riser channel and poured into a stainless steel canister.

Due to similarities in design, most mathematical modeling techniques developed in the commercial glass industry can be directly applied to the waste glass model. The theoretical development of mathematical models in the commercial glass industry has been reviewed in the open literature [3,4,5]. Most of these models solve velocity, temperature, and electric potentials of the glass melt in a furnace with glass properties such as color, viscosity, electric resistivity, and density as functions of temperature. Large scale finite element and finite difference models involved furnace operational parameters such as minimum residence time, melting index, fining index, and recirculating ratio. The side-entry batch feed provides material source term and boundary conditions for the glass melt model. Combustion gas and glass melt models are usually treated separately. However, due to the substantial mass and heat transfer between glass melt, batch-feed pile, and combustion gas, these models need to be integrated to form a closed system.

The waste glass melter models can be catagorized similarly. The major difference in modeling strategies between commercial glass and waste glass processes is derived from the way feeds enter the melter e.g. side(or top)-entry dry batch feed for the commercial glass furnace as opposed to top-entry wet slurry feed for the waste glass melter. The cold cap is an entity unique to the waste glass melter and creates highly unstable velocity and temperature field. From modeling point of view, this cold cap is the major source of computational difficulties that are encountered in defining modeling domain, prescribing boundary conditions, and obtaining stable converging solutions.

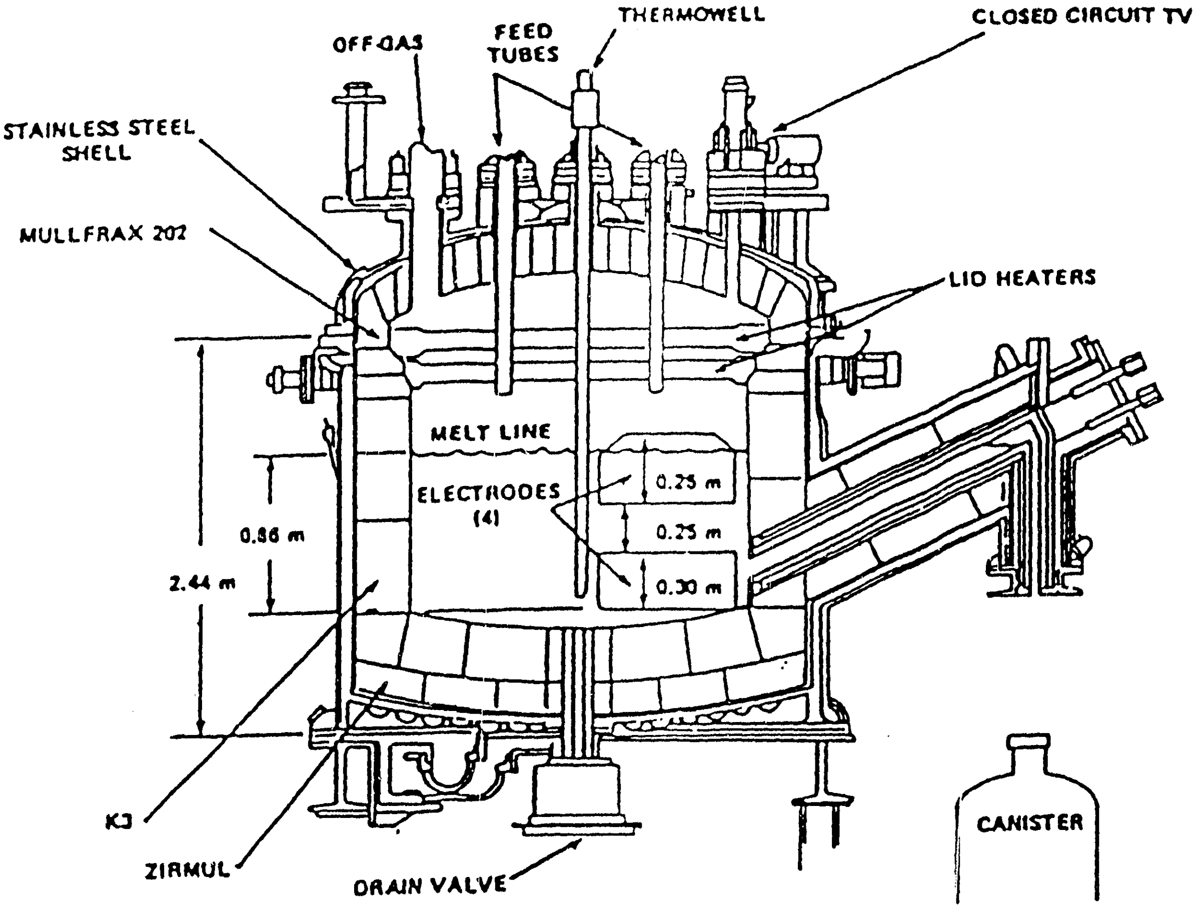

Figure 1. Cross Section of Defense Waste Processing Facility Melter

Melt Pool Convection Model

The melt pool convection model solves 3-D, transient, momentum, energy, and electric equations simultaneously with temperature dependent glass properties as shown in equations (1) through (4).

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \bar{u}) = 0 \tag{1}$$

$$\rho \left(\frac{\partial \bar{u}}{\partial t} + \bar{u} \cdot \nabla \bar{u} \right) = -\nabla P + \nabla \cdot (\mu \nabla \bar{u}) + \rho \bar{g} \{ 1 - \beta (T - T_o) \} \tag{2}$$

$$\rho \cdot C_p \left(\frac{\partial T}{\partial t} + \bar{u} \nabla T \right) = \nabla \cdot (k_T \nabla T) + \nabla E \cdot (k_E \nabla E) \tag{3}$$

$$C_E \frac{\partial E}{\partial t} = \nabla \cdot (k_E \nabla E) + I + S_1 (E) + S_2 (Q) \tag{4}$$

The electric conductivity, K_E,for the waste glass is in the range 0.3 - 0.5 (1/ohm-cm) hence the external magnetic force of induction term was neglected in equation (2). The alternating electric current in the glass melter is quasi-stationary with the frequency in the range 60 - 80 Hz. Therefore the displacement current density $S_1(E)$ can be neglected. The electric charge density due to the movement of glass, $S_2(Q)$, is also small compared to the unit length current density generated in the melt pool by the electrodes. The wave length corresponding to this frequency in a glass melt in the temperature range 1050 - 1150°C exceeds the characteristic length of a melter. Thus the electric field in the glass melt can be regarded as a potential field without eddies.

The energy equation is coupled to the momentum equation through advection and natural convection term in equation (2). The electric equation is coupled to the energy equation through a heat source term which appears as the last term in equation (3). The stiff nonlinearities in these equations require an elaborate iteration scheme for convergence. The hydrothermal instability in the region under the cold cap requires transient simulation approach to obtain a quasi-steady state solution. A typical quasi-steady state for a 2-D one-electrode model three minutes after startup is shown in Figure 2. The breakup of the symmetric flow pattern is clearly shown in this streamline plot.

For a 3-D melter transient model, the melt pool condition can exceed a thermally-induced hydrodynamic stability limit depending on boundary conditions imposed on the model. In this case, dual velocity solutions can exist at fixed geometric locations. An example of such solutions is shown in Figure 3. In this figure, the cutting plane between two electrodes were given two set of nodes. The solutions are exchanged back and forth between the two set of nodes by a small perturbation.

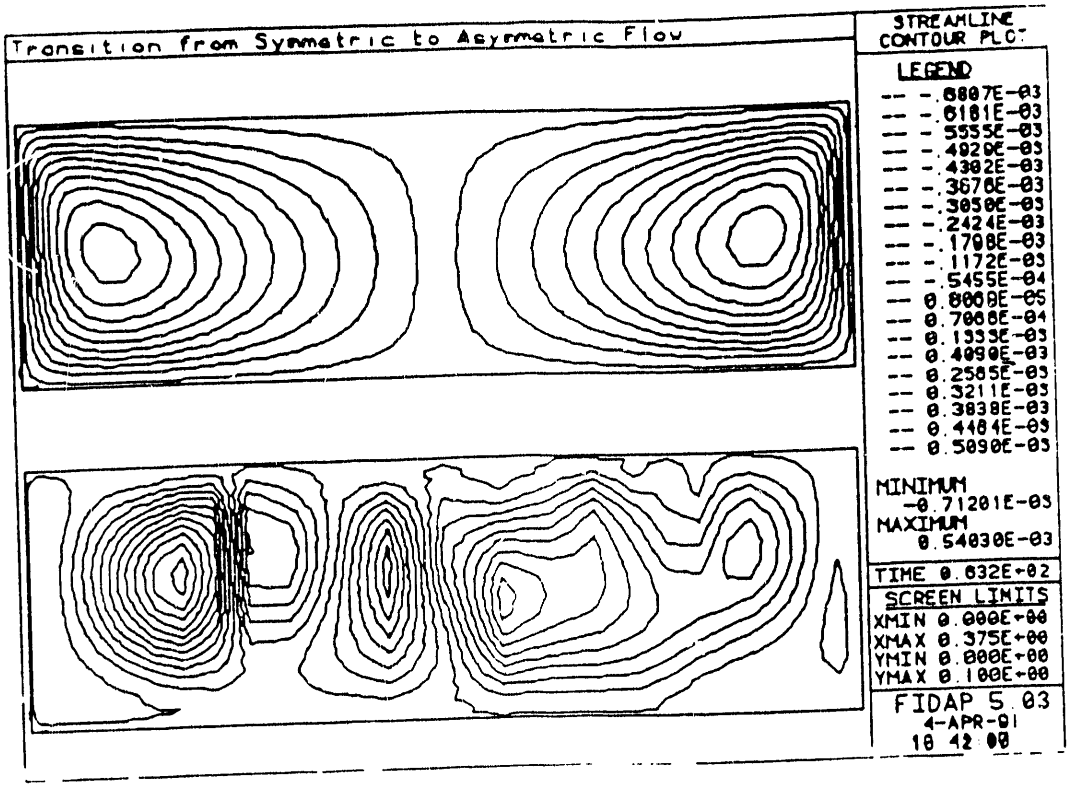

Figure 2. Stream Lines showing Changes of Initial Symmetric Flow Pattern ime (Top) to Quasi-Steady Asymmetric Flow Pattern (Bottom)

A possible form of solutions is a standing wave formed by two waves that have the same amplitude, frequency, but different phase angle propogating in the opposite direction. In normal melter operating condition, the flow can be more stable mainly due to the reduced temperature gradients and enhanced mixing in the melt pool. However, the oscillating solutions produced by hydrodynamic instability are still expected to exist under the cold cap.

In addition to the thermally-induced hydrodynamic instability, the electric field in the melt pool can create an electric instability. The electric instability limit [6]

$$\frac{1}{\rho_E}\frac{\partial \rho_E}{\partial T} \geq 0.3\ (/^{\circ}C) \tag{5}$$

develops a positive feedback of electric currents that can result in electrode melting. The melt pool convection model is valid only if the electric resistivity, ρ_E, stays within this limit.

The choice of a joule heating method for waste glass processing has a significant advantage over direct heating methods. Unlike commercial glasse counterpart, the waste glass has high FeO and FeO_2 contents that make the glass temperature uniformity much harder to achieve. Therefore joule heating and convection current control is the key operational strategy for improving glass quality in waste glass processing.

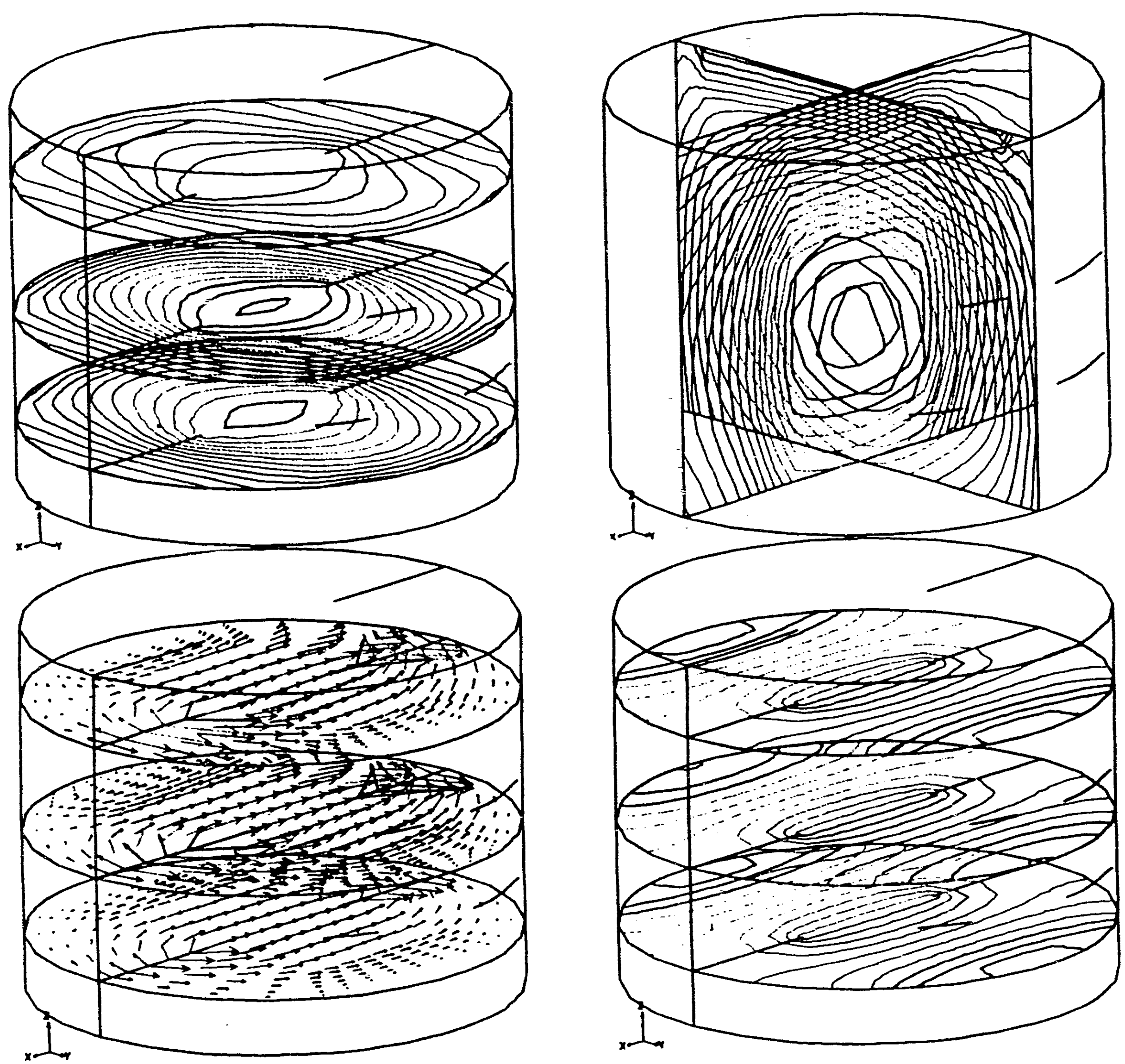

Figure 3. Melter Pool Convection Instability ; Top Left and Right : Temperatures (Max.=1150 °C at Center, Min.=700°C at Top), Bottom Left : Velocity (Max.=2.1 in./sec at Center), Bottom Right : Electric Potentials (Max.=150 Volts at Left, Min.=-150 Volts at Right)

Cold Cap Model

The chemical reactions that take place in the cold cap are difficult to model as there are more than forty different chemical species in a typical waste melter feed composition; thus, Arrhenius type of equations may not be applicable. These chemical reactions produce gases along with reaction heat. The amount of mass transfer of volatile and foaming gases within and through the cold cap is small and may be neglected. However, the effects of these gases on cold cap heat balance can be substantial.

In commercial glass industry, several batch modeling studies [7,8] were conducted wherein the batch was dry, horizontally charged. The melting characteristics were determined by batch geometry and charging speed. In the waste melter, however, the cold cap's melt rate and its movement when it melts at the bottom interface with the molten glass is small. Therefore, a first-order cold cap model can be developed with an overall melter system energy balance alone.

Boundary conditions of a cold cap model are not clearly defined. The boundaries include radiation from plenum heaters, convection by steam and gases, and melting layer diffusion and convection at the bottom. The primary requirement in developing an acceptable cold cap model is to determine these boundaries and boundary conditions. As the cold cap comprises a combination of solid, liquid, and gaseous phases in series, it might be treated as a composite material with different conductivities for a thermal model.

The plenum model is closely linked to the cold cap model but may be treated independently. The plenum models involve radiation exchange through participating media, turbulence, and combustion. The radiation participating media comprise steam and gases that emit and absorb radiation. The models for radiation, combustion, and turbulence should be coupled and solved simultaneously. Slurry feed rate, plenum heater temperature, and steam enthalpy are the key parameters for these models.

Noble Metal Deposit Model

The noble metal deposit process is closely coupled with cold cap diffusion process. It is not well known how noble metals are released form the cold cap. During the solid-to-liquid phase changes which may be treated as a Stephan type problem [9], glass enthalpy changes and natural convection takes place at the solid-to-liquid interface. Assuming that such an interface exists, the noble metals might be assumed to be released at that interface. Therefore the extent of the glass-melt model boundary needs to be defined for a noble metal deposit model.

Assuming that noble metal particle diffusion is negligible, the conservation equation for the number of noble metal particles per unit volume and per unit size interval can be expressed as:

$$\frac{\partial}{\partial x}(\rho u m) + \frac{\partial}{\partial y}(\rho v m) + \frac{\partial}{\partial z}[\rho(w - w_p)m] = -M\frac{\partial}{\partial s}(\dot{S}\frac{\rho m}{M}) \tag{6}$$

where u,v, and w represent glass velocities and w_p is the relative velocity of noble metal particles in the vertical direction. M is the mass of a single particle, m denotes mass fraction of grains per unit size interval, s is the size of the particle, and S is the rate of change of particle size which can also be interpreted as velocity of the particle in geometrical size space.

In noble metal deposit process, the particles amalgamate more than diffuse. As a first order approximation, the amalgamation process may be expressed as an exact reversal of diffusion process as following.

$$\dot{S} = \frac{ds}{dt} = D\,(C_\infty - C_i)/\rho s \tag{7}$$

The geometrical size is characterized by the particle radius, C_∞ is the noble metal concentration in the bulk glass, and C_i is the liquid-grain interface at equilibrium. If the range of seed size interval is divided into a finite set of intervals, the equation (5) can be used for each interval to determine size distribution.

Assuming that noble metal is spherical in shape, its settling velocity in the presense of natural convection field can be expressed as

$$w_p = \frac{2\,s^2}{9\,\mu}\,(\rho_m - \rho_g\,(1 + \beta\,\Delta T))\,g \tag{8}$$

where subscript m and g denotes noble metal and glass respectively.

A marginal particle size may be defined as the particle size which has a settling velocity equal to the average convection current velocity for a given melter operating condition. If the particle diameter exceeds this marginal diameter, it will settle on the melter floor. This marginal diameter, D_e ,can be expressed as

$$D_e = \sqrt{\frac{18\,\mu_g w_p}{g\,(\rho_m - \rho_g)\,(1 - \beta\,\Delta T)}} \tag{9}$$

The residence time is defined as the time single noble metal particle travels before it encounters another particle and amalgamates. The residence time may be expressed as

$$t_r = \frac{D_e^2 A_{avg}}{8 \int_A \frac{D(C_i - C_\infty)}{\rho_g} dA} \qquad (10)$$

where A_{avg} is cross sectional area of primary convection current tube. The primary convection current is defined as the convection current generated by the sidewall and floor heat loss whereas the secondary convection current is the unstable convection current generated under the cold cap as shown in Figure 3. In normal operating condition, four symmetric primary convection cells are coupled with smaller secondary oscillating convection cells above are expected to be present. Then the primary convection current tube would be approximately one-quater of the melter diameter.

The numerical techniques developed in commercial glass industry to simulate silica batch grains or gas bubble distributions [10,11,12,13] can be modified to address some of the issues in the noble metal deposit problem. These models predict grain size distribution, bubble concentration, trajectories, and particle chemical reaction [10].

CONCLUSIONS

Technical issues to be encountered in the operation of DWPF melter at Savannah River Site were identified and mathematical modeling techniques required to address these issues were discussed. The mathematical models can be catagorized in three broad areas: melt pool convection model, cold cap model, and noble metal deposit model. The melt pool convection model involves solving a system of transient momentum, energy, and electric equations with temperature dependent properties. Due to the hydrothermal instability inherent to liquid slurry feed type melter, a full 3-D model is required to capture asymmetric thermal and flow profiles. At the interface between the melt pool and cold cap where hydrothermal stability can be exceeded, multiple velocity solutions can exist due to instability-induced bifurcation.

The cold cap model has been developed in the past mostly for chemical reactions of vitrifying chemical species. However, modeling efforts on the hydrothermal process in the cold cap are far from being complete due to the complexity of the underlying physical phenomena. The noble metal deposit problem is the most critical waste melter operational issue to date. Some of the ongoing efforts involve prediction of noble metal size distribution, particle tracking, amalgamation, settling velocity, and deposit rate. Due to limitations on in-situ experiments that can be performed on a radioactive waste melter, application of modeling techniques as alternative numerical experiment tools to resolve these issues has become more crucial than ever.

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